Synthetic incoherence for electron microscopy

Zachary H. Levine[1,*] and Robyn M. Dunstan[1,2]

[1]*National Institute of Standards and Technology, Gaithersburg, Maryland 20899-8410, USA*
[2]*Elizabethtown College, Elizabethtown, Pennsylvania 17022-2298, USA*
**Corresponding author: zlevine@nist.gov*

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Tomographic studies of submicrometer samples in materials science using electron microscopy have been inhibited by diffraction effects. In the present work, we describe a practical method for ameliorating these effects. First, we find an analytic expression for the mutual coherence function for hollow-cone illumination. Then, we use this analytic expression to propose a Gaussian weighting of hollow-cone illumination, which we name tapered solid-cone illumination, and present an analytic expression for its mutual coherence function. Finally, we investigate numerically an n-ring approximation to tapered solid-cone illumination. The results suggest a method for removing diffraction effects and hence enabling tomography. © 2007 Optical Society of America

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1. INTRODUCTION

Mainstream tomography requires an incoherent signal [1]. However, the source of an electron microscope is highly coherent. In biology, the most common solution is to ensure that the sample is amorphous [2]. Plunge freezing prevents the formation of ice crystals; otherwise, the complexity of a biological cell ensures the sample is amorphous. However, in materials science, small-scale crystals are extremely common. Crewe and co-workers introduced a method for incoherent imaging in the scanning transmission electron microscope (STEM) using a high-angle annular dark field (HAADF) detector [3]. The method is also known as Z-contrast imaging; indeed Crewe and co-workers imaged a single uranium atom on a carbon film. The method was applied to tomography by Midgley and co-workers [4]. For high-angle scattering, incoherence is achieved because the scattering is primarily due to phonons; formally, the Debye–Waller factor is very small. Less formally, if k is the wave vector describing the scattering and $\bar{u}$ is the rms deviation of the atoms about their lattice positions, whenever $k\bar{u} \gg 1$, there is no average phase relation between the scattering of an atom and its neighbor, so the sample is effectively amorphous. The complement of the ideal HAADF signal has also been introduced, under the name of incoherent bright field (IBF) imaging [5], with the additional advantage that the signal is a monotonic function of thickness, whereas a practical HAADF detector (with an inner diameter and an outer diameter) has a single-peaked thickness-intensity relation. A disadvantage to the HAADF or IBF approach is that relatively few electrons (typically 1%) are scattered into or out of the detector, respectively.

Recently, Levine [6] proposed a combination scanning and imaging mode to create an incoherent beam on the sample. The proposal called for scanning with a dosage given by a Gaussian. Theoretically, the nonnegligible region of the mutual coherence function can be confined to a Gaussian with a standard deviation well below 1 nm. Of course, hollow-cone illumination has long been used to reduce the coherence in imaging [7,8]. Large-angle hollow-cone illumination [9] represents an alternative to HAADF to achieve Z-contrast imaging in which part of the incoherence is achieved as in conventional HAADF and part is due to the angular averaging of hollow-cone imaging.

The purpose of the present paper is threefold: first, to present analytic results for the mutual coherence function for hollow-cone illumination; second, to use these analytic results to propose another illumination scheme, named here tapered solid-cone illumination, and present its mutual coherence function; and finally, to address the merits of an n-ring (i.e., n-hollow cone) approximation to tapered solid-cone illumination, which may be implemented with hollow-cone illumination using a commercial TEM. As in earlier work [6,7], we demonstrate here an alternative to hollow-cone illumination, which leads to the nonnegilgible region of the mutual coherence function to be more localized.

The definition of the mutual coherence function is given in many sources, such as Spence [10]:

$$\Gamma(\vec{r}_1,\vec{r}_2,T) = \lim_{\tau\to\infty} \frac{1}{2\tau}\int_{-\tau}^{\tau} \mathrm{d}t\, \psi^*(\vec{r}_1,t)\psi(\vec{r}_2,t+T), \tag{1}$$

where $\vec{r}_1$ and $\vec{r}_2$ are two positions in the optical field ψ, t is a time, and T is a time difference. Because the source is nearly monochromatic, only the equal time case, i.e., $T=0$, is of interest here [11]. The complex degree of coherence is defined as the normalized mutual coherence function:

$$\gamma(\vec{r}_1,\vec{r}_2) = \frac{\Gamma(\vec{r}_1,\vec{r}_2)}{[\Gamma(\vec{r}_1,\vec{r}_1)\Gamma(\vec{r}_2,\vec{r}_2)]^{1/2}}. \tag{2}$$

The normalized mutual coherence function is an intensity-weighted average of the phase factor between two illuminated points. When $|\gamma(\vec{r}_1,\vec{r}_2)| \ll 1$, the beam is incoherent. Of course, $\gamma(\vec{r},\vec{r})=1$, so the best one can do is to have $|\gamma(\vec{r}_1,\vec{r}_2)|$ go to zero rapidly as $|\vec{r}_1-\vec{r}_2|$ grows.

2. GAUSSIAN BEAMS

We assume that the beam is a Gaussian solution of the paraxial ray equation of scalar diffraction theory. Actual beams in electron microscopes may be better described by a Gaussian times a Hermite polynomial, which is also a solution to the paraxial ray equation, but we ignore this complication here.

The Gaussian solution to the paraxial ray approximation for a beam traveling with its center in the z' direction is [12]

$$\psi(x',y',z') = \frac{1}{1+iZ}\exp\left(-\frac{X^2+Y^2}{1+iZ}\right), \tag{3}$$

where the scaled dimensions X, Y, and Z are related to the physical dimensions x', y', and z' by

$$X = x'/r_0,$$
$$Y = y'/r_0,$$
$$Z = \lambda z'/\pi r_0^2 = z'/l_R, \tag{4}$$

where r_0 is a parameter characteristic of the beam waist corresponding to two standard deviations of the intensity, i.e., the square of the wave function; λ is the wavelength; and the Rayleigh length l_R is defined implicitly. Primes are used to reserve x, y, and z for the frame of the sample. The beam waist may be given in terms of the angle θ_1 between the direction of maximum intensity and the angle at which the intensity has fallen by a factor of e^2:

$$r_0 = \lambda/\pi\theta_1. \tag{5}$$

In Eq. (1), we are led to consider a time average of the illumination. Although this time average arises commonly from a statistical source, such as thermal effects in a light bulb, the definition also admits a controlled variation of the illumination. The paper is an exploration of the mutual coherence function for various controlled variations of the illumination, a procedure we call "synthetic incoherence." Abstractly, the beams we consider may be described in a five-parameter space consisting of the three-dimensional (3D) position of the beam center relative to the sample and the direction (a 2D point on the unit sphere) of the beam relative to the z axis. The various sections below may all be viewed as particular averages in this five-parameter space. (In this paper and an earlier one [6], the averages are over no more than two parameters.)

3. HOLLOW-CONE ILLUMINATION

In hollow-cone illumination, the focal point of the beam is common throughout the averaging process. This point is taken as the origin. The center of the sample is taken to lie at a point $(0,0,z)$, i.e., directly under the focal spot. The half-angle of the hollow cone is taken to be θ. The averaging of hollow-cone illumination consists of varying the azimuthal angle φ. Formally, the beam is related to the laboratory frame by the orthogonal transformation

$$\begin{pmatrix} x' \\ y' \\ z' \end{pmatrix} = \begin{bmatrix} \cos\theta\cos\varphi & \cos\theta\sin\varphi & -\sin\theta \\ -\sin\varphi & \cos\varphi & 0 \\ \sin\theta\cos\varphi & \sin\theta\sin\varphi & \cos\theta \end{bmatrix} \begin{pmatrix} x \\ y \\ z \end{pmatrix}. \tag{6}$$

The variables x', y', and z' are related to the dimensionless variables X, Y, and Z by Eq. (4). If we make the small-angle approximation $\theta \ll 1$ and the related approximation $l_R^{-1} \ll r_0^{-1}$, both well justified for an electron microscope, the dimensionless coordinates are given approximately from the laboratory coordinates by

$$\begin{pmatrix} X \\ Y \\ Z \end{pmatrix} \approx \begin{bmatrix} \cos\varphi/r_0 & \sin\varphi/r_0 & -\theta/r_0 \\ -\sin\varphi/r_0 & \cos\varphi/r_0 & 0 \\ 0 & 0 & 1/l_R \end{bmatrix} \begin{pmatrix} x \\ y \\ z \end{pmatrix}. \tag{7}$$

The mutual coherence function for hollow-cone illumination is given in the average over φ of a Gaussian beam tilted at fixed θ evaluated at two points $\vec{r}_1$ and $\vec{r}_2$ where each vector has Cartesian components given by $\vec{r}_j = x_j\hat{x} + y_j\hat{y} + z_j\hat{z}$:

$$\Gamma_{HC}(\vec{r}_1,\vec{r}_2) = \frac{1}{2\pi}\int_0^{2\pi} d\varphi \prod_{j=1}^{2}\left(1 \mp i\frac{z_j}{l_R}\right)^{-1} \times \exp\left(-\frac{x_j^2+y_j^2-2\theta z_j(x_j\cos\varphi + y_j\sin\varphi)+\theta^2 z_j^2}{r_0^2(1\mp i z_j/l_R)}\right). \tag{8}$$

We introduce the convention that the top sign is used for $j=1$ and the bottom sign for $j=2$.

To perform the integral, we introduce the variables

$$\alpha = \frac{2\theta}{r_0^2}\sum_{j=1}^{2}\frac{z_j x_j}{i \pm z_j/l_R},$$
$$\beta = \frac{2\theta}{r_0^2}\sum_{j=1}^{2}\frac{z_j y_j}{i \pm z_j/l_R}. \tag{9}$$

These may be further transformed via

$$\alpha = \Omega\cos\psi,$$
$$\beta = \Omega\sin\psi. \tag{10}$$

Equation (10) implies that

$$\alpha^2+\beta^2 = \Omega^2, \tag{11}$$

even though α, β, Ω, and ψ are all complex. Omitting factors independent of φ, the integral of Eq. (8) may be written as

$$I = \int_0^{2\pi} \exp[i\Omega(\cos\psi\cos\varphi + \sin\psi\sin\varphi)] = \int_0^{2\pi}\exp[i\Omega\cos(\varphi-\psi)] = 2\pi J_0(\Omega). \tag{12}$$

Since $J_0(\Omega)$ is an even function, the answer does not depend on the sign of the square root implicit in finding Ω from Eq. (11). In passing from the second to the third lines above, we may change variables to some $\bar{\varphi}=\varphi-\psi$ and use Cauchy's theorem to deform the contour of integration over $\bar{\varphi}$ into three parts: from $-\psi$ to 0, from 0 to 2π, and from 2π to $2\pi-\psi$. The first and third parts cancel because of the periodicity of the integrand, and the second is Poisson's integral [13]. Writing the solution in the original variables, we have

$$\Gamma_{HC}(\vec{r}_1,\vec{r}_2) = \left[\prod_{j=1}^{2} \left(1 \mp i \frac{z_j}{l_R}\right)^{-1} \exp\left(-\frac{x_j^2+y_j^2+\theta^2 z_j^2}{r_0^2(1\mp i z_j/l_R)}\right)\right] J_0(\Omega), \tag{13}$$

with

$$\Omega = \frac{2\theta}{r_0^2}\left[\left(\sum_{j=1}^{2}\frac{z_j x_j}{i \pm z_j/l_R}\right)^2 + \left(\sum_{j=1}^{2}\frac{z_j y_j}{i\pm z_j/l_R}\right)^2\right]^{1/2}. \tag{14}$$

To gain some insight into the relevant values of Ω, consider the special case of $z=z_1=z_2$ and $y_1=y_2=0$. Then the phase $\arg\Omega=\tan^{-1}([(x_2+x_1)l_R]/[(x_2-x_1)z])$ may be estimated. Typically, $x_2+x_1\sim\theta_1 z$; so, using Eqs. (4) and (5), a substantially real value will be achieved whenever $|x_2-x_1|\gg\lambda/(\pi\theta_1)$. In our typical case, $\lambda=1.969$ pm and $\theta_1=3$ mrad, so crossover to real behavior would occur at a separation of 209 pm. For the purposes of estimation, if not calculation, the argument of the Bessel function may be taken to be real for separations in the range of interest, i.e., above 1 nm for tomography.

The special case of Eq. (13) required for the denominator of the normalized mutual coherence function is

$$\Gamma_{HC}(\vec{r}_j,\vec{r}_j) = \left(1+\frac{z_j^2}{l_R^2}\right)^{-1} \times \exp\left(-2\frac{x_j^2+y_j^2+\theta^2 z_j^2}{r_0^2[1+(z_j/l_R)^2]}\right) I_0\left(\frac{4\theta z_j (x_j^2+y_j^2)^{1/2}}{r_0^2\,[1+(z_j/l_R)^2]}\right), \tag{15}$$

which contains the modified Bessel function I_0, which is nonoscillatory because its argument is real.

Finally, we are in a position to write an approximation to the normalized mutual coherence function for hollow-cone illumination:

$$\gamma_{HC}(\vec{r}_1,\vec{r}_2) = \left\{\prod_{j=1}^{2}\exp\left[\pm i\tan^{-1}\left(\frac{z_j}{l_R}\right) \mp i\frac{x_j^2+y_j^2+\theta^2 z_j^2}{r_0^2}\frac{z_j/l_R}{1+(z_j/l_R)^2}\right]\right\} \times J_0(\Omega)\left[\prod_{j=1}^{2} I_0\left(\frac{4\theta z_j(x_j^2+y_j^2)^{1/2}}{r_0^2\,[1+(z_j/l_R)^2]}\right)\right]^{-1/2}. \tag{16}$$

The numerical approximations we have made appear to be very modest. An example of the normalized mutual coherence function is given in Fig. 1. A fully numerical calculation using the exact transformations is compared with the results from Eqs. (13) and (15). Because the results are so close, in order to compare them it was necessary to plot the difference. We also show the difference of the Bessel function to its asymptotic formula [14],

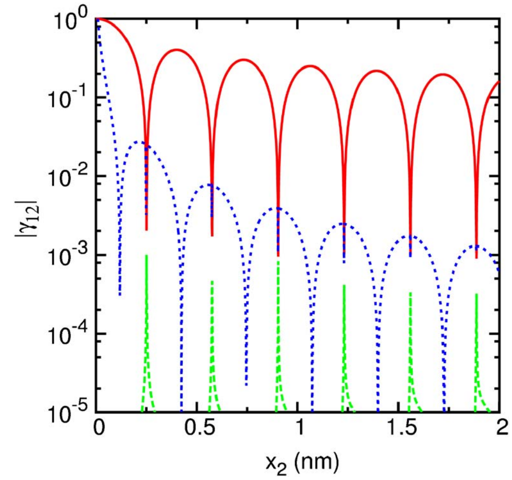

Fig. 1. (Color online) Absolute value of the normalized mutual coherence function for hollow-cone illumination calculated numerically (solid curve), absolute value of the difference between that and the analytic solution of Eq. (13) (dashed bottom curve), and the absolute value of the difference between the asymptotic expression of Eq. (17) and the analytic solution (dotted middle curve). The parameters are $\theta=3$ mrad, $\theta_1=3$ mrad, $\lambda=1.969$ pm (corresponding to a 300 keV electron energy), $z_1=z_2=1$ mm, and $x_1=y_1=y_2=0$.

$$J_0(z)\approx\left(\frac{2}{\pi z}\right)^{1/2}\cos\left(z-\frac{\pi}{4}\right), \tag{17}$$

which shows that the asymptotic form may be used for qualitative understanding.

4. TAPERED SOLID-CONE ILLUMINATION

It was shown in an earlier paper that the normalized mutual coherence function could go to zero like a Gaussian if the illumination conditions were suitably arranged [6]. Here, we find another method for doing so based on what we call tapered solid-cone illumination. Specifically, we will average the hollow-cone illumination over the parameter θ with the function

$$W(\theta)=\frac{\theta}{\theta_0^2}\exp\left(-\frac{\theta^2}{2\theta_0^2}\right), \tag{18}$$

where θ_0 is a constant (chosen to be the standard deviation of the Gaussian) and $\int_0^\infty d\theta W(\theta)=1$. The linear θ weighting may be viewed as the Jacobian in polar coordinates, so there is a uniform illumination per unit area for $\theta\ll\theta_0$. Because we anticipate θ_0 to be in the milliradian range, there is very little approximation by the replacement of the maximum value of θ, namely π, by ∞.

If Eq. (13) is weighted with Eq. (18), we are led to consider an integral

$$I=\int_0^\infty d\theta\,\theta\exp\left(-\frac{\theta^2}{2\theta_2^2}\right)J_0(\omega\theta), \tag{19}$$

where $\omega\theta=\Omega$ with Ω given by Eq. (14) and

$$\theta_2^{-2} = \theta_0^{-2} + \frac{2}{r_0^2}\sum_{j=1}^{2} \frac{z_j^2}{1 \mp i z_j/l_R}. \tag{20}$$

Using the tabulated integral [15]

$$\int_0^\infty d\theta\; \theta \exp(-a\theta^2) J_0(b\theta) = \frac{1}{2a}\exp\left(-\frac{b^2}{4a}\right), \tag{21}$$

we obtain

$$\Gamma_{TC}(\vec{r}_1,\vec{r}_2) = \left[\prod_{j=1}^{2}\left(1 \mp i\frac{z_j}{l_R}\right)^{-1} \times \exp\left(-\frac{x_j^2+y_j^2}{r_0^2(1 \mp i z_j/l_R)}\right)\right]\frac{\theta_2^2}{\theta_0^2}\exp\left(-\frac{\theta_2^2\omega^2}{2}\right). \tag{22}$$

For the cases we consider here, θ_2 is comparable to θ_0. Also, if $z_1=z_2$, then θ_2 is real.

To obtain the normalized mutual coherence function, the denominator requires the special case of Eq. (22):

$$\Gamma_{TC}(\vec{r}_j,\vec{r}_j) = \left(1+\frac{z_j^2}{l_R^2}\right)^{-1}\exp\left(-\frac{2}{r_0^2}\frac{x_j^2+y_j^2}{1+(z_j/l_R)^2}\right)\frac{\theta_{2j}^2}{\theta_0^2} \times \exp\left(\frac{8\theta_{2j}^2}{r_0^4}\frac{z_j^2(x_j^2+y_j^2)}{[1+(z_j/l_R)^2]^2}\right), \tag{23}$$

where

$$\theta_{2j}^{-2} = \theta_0^{-2} + \frac{4}{r_0^2}\frac{z_j^2}{1+(z_j/l_R)^2}. \tag{24}$$

The normalized mutual coherence function is given by

$$\gamma_{TC}(\vec{r}_1,\vec{r}_2) = \left\{\prod_{j=1}^{2}\frac{\theta_2}{\theta_{2j}}\exp\left[\pm i\tan^{-1}\left(\frac{z_j}{l_R}\right) \mp i\frac{x_j^2+y_j^2}{r_0^2}\frac{z_j/l_R}{1+(z_j/l_R)^2}\right]\right\} \times \exp\left(-\frac{\theta_2^2\omega^2}{2} - \sum_{j=1}^{2}\frac{4\theta_{2j}^2}{r_0^4}\frac{z_j^2(x_j^2+y_j^2)}{[1+(z_j/l_R)^2]^2}\right). \tag{25}$$

Hence, tapered solid-cone illumination causes the normalized mutual coherence function to vanish like a Gaussian in the separation of the two positions, which are its arguments.

Next, we give a representative example to illustrate the typical magnitudes. We consider only $|\gamma_{TC}|$. For the parameters $z=z_1=z_2=1$ mm, hence $\theta_2=\theta_{2j}$, $\lambda=1.969$ pm, $\theta_0=\theta_1=3$ mrad, and $y_1=y_2=0$, if we vary x_1-x_2 holding x_1+x_2 constant, then, using Eqs. (14) and (25) and neglecting 1 compared with z/l_R,

$$|\gamma_{TC}| \sim \exp\left(-2\theta_2^2\frac{l_R^2}{r_0^4}(x_1-x_2)^2\right) = \exp\left[-\left(\frac{2^{1/2}\pi\theta_2}{\lambda}\right)^2\frac{(x_1-x_2)^2}{2}\right]. \tag{26}$$

Under these assumptions, $|\gamma_{TC}|$ decreases like a Gaussian with a standard deviation of $\lambda/(\sqrt{2}\pi\theta_2)=210$ pm. This result suggests that essentially complete incoherence may be achieved in less than 1 nm using practical choices for all parameters. The illumination conditions proposed are therefore suitable for enabling incoherent illumination for tomography.

Typically, the vendor supports hollow-cone illumination but not tapered solid-cone illumination. Hence, we consider discrete approximations of tapered solid-cone illumination by a finite set of hollow-cone illuminations, called here the n-ring approximation. We consider the approximation of Eq. (21) by a weighted sum of discrete values of θ. If we introduce $u=\theta^2/(2\theta_2^2)$, Eq. (19) becomes

$$I = \theta_2^2\int_0^\infty du\; J_0(\sqrt{2}\theta_2\omega\sqrt{u})\exp(-u). \tag{27}$$

In this form, the highly efficient Laguerre integration [16] may be applied. The results for the normalized mutual coherence function averaged over two, four, and eight hollow cones with θ used to emulate the exact tapered-cone illumination are shown in Fig. 2 along with the results for tapered solid-cone illumination and hollow-cone illumination, which may be regarded as a one-point approximation to the integral.

The results may be summarized by the statement that the n-ring approximation represents most of the first n standard deviations adequately; in more detail, the relation appears to be sublinear. Afterward, the results diverge and revert to an envelope given by the asymptotic form of Eq. (17). With care, it appears to be possible to obtain good incoherence on medium scales. Such illumination may be suitable for tomography; however, tapered solid-cone illumination emulated by a few values of hollow-cone illumination is not a fully satisfactory substitute for properly implemented tapered solid-cone illumination because of coherence at long length scales. The key advantage of the n-term approximation is that it may be implemented on an existing TEM merely by running hollow-cone illumination many times with suitable angles and taking a weighted average of the results, whereas

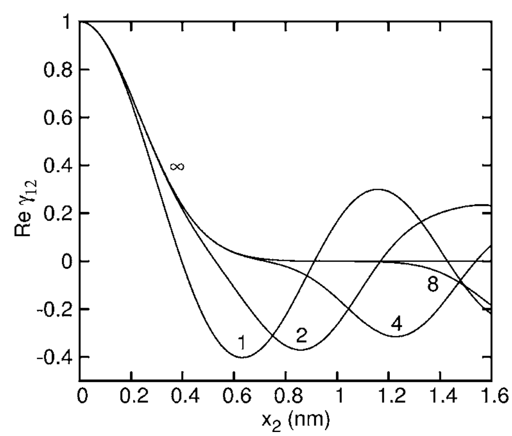

Fig. 2. Real part of normalized mutual coherence function for the average of one, two, four, and eight hollow cones using Laguerre integration points and weights [16] and the analytic limit (labeled ∞), given in Eq. (25). The imaginary part is very small. Parameters are given in the text. For small x_2, not all of the curves are distinguishable.

implementing true tapered-cone illumination will probably require cooperation of a vendor.

5. DISCUSSION AND CONCLUSIONS

Standard tomography is based on projections, i.e., beams propagating in a straight line from the entrance to the exit. However, an incoherent beam must come from a range of directions. These conditions are compatible if the divergence of the incoherent beam is not too large and the number of voxels across the sample is also not too large. Specifically, the edge of validity for the projection approximation occurs if two beams entering the sample at a given voxel emerge at adjacent voxels. If θ is the divergence angle of the beam and N is the sample thickness in voxels, we require $\theta < N^{-1}$ for the projection assumption to be valid. To optimize the operating conditions, the standard deviation of the normalized mutual coherence function for tapered solid-cone illumination should be chosen to be as large as reasonably possible (i.e., just a little smaller than the pixel width) so that the beam is as parallel as possible to allow many voxels. For large systems or the highest resolution, ultimately the projection assumption will need to be abandoned.

Incoherent illumination in TEM may enable tomography of small crystalline samples with a dose that is lower than previously possible using HAADF by 1 to 2 orders of magnitude. Tapered solid-cone illumination is the most promising scheme presently available because it leads to a very rapid decrease in the mutual coherence, has no appreciable long-range tail, and may be implemented with limited change to the existing hardware and software of a modern commercial transmission electron microscope. Specifically, one needs to add a good method for averaging over the tilt of the cone angle in hollow-cone illumination.

Because we have made use of only scalar wave theory, the results could be applied to other areas of optics, e.g., achieving incoherent illumination with laser beams or synchrotron undulator radiation. However, a small numerical aperture is required for the validity of our approximations.

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